PATENT ABSTRACTS OF JAPAN

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(54) ELASTIC SEALING MATERIAL

(57)Abstract:

PROBLEM TO BE SOLVED: To provide an elastic sealing material of a one-component type acrylic emulsion type, capable of improving elastic recovery without reducing other properties such as elongation.

SOLUTION: This elastic sealing material comprises a copolymer composed of (a) 84.5-98.88 wt.% acrylic ester having a 4-12C alkyl group, (b) 1-10 wt.% acrylonitrile and/or methacrylonitrile, (c) 0.1-5 wt.% α,β-unsaturated carboxylic acid, and (d) 0.02-0.5 wt.% ethylenically unsaturated polyfunctional crosslinkable monomer.

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* NOTICES *

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] the sealing material with which this invention has elasticity -- it is related with an acrylic emulsion mold elastic sealing compound in detail.
[0002]
[Description of the Prior Art] A structural sealing material is divided roughly into an indeterminate form sealing material and a fixed form sealing material. There are an inelastic mold and an elastic mold in the above-mentioned indeterminate form sealing material. There are glazing putty, an oily caulking material, an asphalt system, etc. in this inelastic mold, it is divided into one component type and two component types by the above-mentioned elastic mold according to a gestalt, and one component type is divided into a non-solvent mold, a solvent mold, and an emulsion mold by ** and also the hardening device. This emulsion mold is the same desiccation hardening mold as a solvent mold, and has acrylic and a styrene butadiene system.
[0003] This acrylic emulsion mold elastic sealing compound is piping joint, such as surface treatment for waterproofing, such as crack repair of concrete, an ALC plate, a block, etc., an extractives pan JON joint, the joint of the part which receives vibration, repair of a crack, a steel structure, an ALC joint, a waterworks, gas, wastewater, and electrical and electric equipment, and a sealing material used for a duct joint list by repair of an outer wall crack etc.
[0004] Since this acrylic emulsion mold elastic sealing compound is aquosity, it is no odor and easy handling.
[0005]
[Problem(s) to be Solved by the Invention] However, this was difficult, although only elastic stability (rubber-elasticity) needed to be raised without this acrylic emulsion mold elastic sealing compound reducing the elongation at the time of the highest reinforcement, and the elongation at the time of fracture in order to use it for the application which needs the elasticity stability of high performance. For this reason, 2 component silicone system elastic sealing compound, 2 component polysulfide system elastic sealing compound, 2 component conversion silicone system elastic sealing compound, 1 component silicone system elastic sealing compound, etc. are used for the application which needs the elastic stability of high performance. However, handling takes each of these cautions while the cure against a smell is needed, since the solvent is used.
[0006] Then, this invention aims at offering the acrylic emulsion mold elastic sealing compound of one component type which raises elastic stability, without reducing other physical properties, such as elongation.
[0007]
[Means for Solving the Problem] As for the artificer of this invention, the carbon number of the (a) alkyl group a sealing material 84.5 - 98.88 % of the weight of acrylic ester of 4-12, (b) Acrylonitrile and/or 1 - 10 % of the weight of methacrylonitriles, (c) Constitute from alpha, 0.1 - 5 % of the weight of beta-unsaturated carboxylic acid, and a copolymer of 0.02 - 0.5 % of the weight of (d) ethylene nature partial

saturation polyfunctional cross-linking monomers. That is, while being able to make elastic stability discover by adding and carrying out copolymerization of the (d) component to each component of above-mentioned (a) - (c), it found out that other physical properties, such as elongation, could be made to hold, and the above-mentioned technical problem was solved.
[0008]
[Embodiment of the Invention] Hereafter, the operation gestalt of this invention is explained.
[0009] The elastic sealing compound concerning this invention is an acrylic emulsion mold elastic sealing compound which consists of a copolymer with which the carbon number of an alkyl group contains alpha and beta-unsaturated carboxylic acid as acrylonitrile and/or a methacrylonitrile, and a (c) component, and contains four components of an ethylene nature partial saturation polyfunctional cross-linking monomer as (d) as the acrylic ester of 4-12, and a (b) component as a (a) component.
[0010] As an example of the above-mentioned (a) component, butyl acrylate, acrylic-acid amyl, 2-ethylhexyl acrylate, acrylic-acid heptyl, acrylic-acid iso octyl, acrylic-acid n-nonyl, acrylic-acid iso nonyl, acrylic-acid DESHIRU, acrylic-acid lauryl, etc. are raised. This (a) component can be used not only combining the one above-mentioned sort but combining two sorts or more. The amount of this (a) component used has 84.5 - 98.88 good % of the weight to the whole copolymer, and its 89.2 - 96.97 % of the weight is desirable. If it is this within the limits, it is possible not to check the property which other copolymerization components show, and to give elasticity, and it is economically advantageous.
[0011] The amount of the above-mentioned (b) component used has 1 - 10 good % of the weight to the whole copolymer, and its 2.5 - 7.5 % of the weight is desirable. If fewer than 1 % of the weight, elasticity will tend to become inadequate, and if [than 10 % of the weight] more, a sealing material may become hard too much.
[0012] As an example of the above-mentioned (c) component, an acrylic acid, a methacrylic acid, a maleic acid, a maleic anhydride, an itaconic acid, a crotonic acid, a fumaric acid, etc. are raised. This (c) component can be used not only combining the one above-mentioned sort but combining two sorts or more. The amount of this (c) component used has 0.1 - 5 good % of the weight to the whole copolymer, and its 0.5 - 3 % of the weight is desirable. If fewer than 0.1 % of the weight, a sealing material's stability will fall, and if [than 5 % of the weight] more, a water resisting property may get worse.
[0013] As an example of the above-mentioned (d) component, there are glycidyl group content monomers, such as divinyl compounds, such as a divinylbenzene, diallyl phthalate, ethylene glycol dimethacrylate, trimethylolpropane triacrylate, and allyl compound acrylate, glycidyl methacrylate, and glycidyl acrylate, N-methylol acrylamide, etc., the amount used has 0.02 - 0.5 good % of the weight to the whole copolymer, and its 0.05 - 0.2 % of the weight is desirable. At less than 0.02 % of the weight, cross-linking is low and the satisfactory effectiveness of elastic stability in this invention is not acquired. Moreover, if 0.5 % of the weight is exceeded, cross-linking will become high too much and will bring about the fall of the elongation engine performance etc.
[0014] As the above-mentioned (d) component, a GURIJIRU radical content monomer is desirable and glycidyl methacrylate is more desirable.
[0015] The glass transition temperature (it abbreviates to "Tg" hereafter.) of the above-mentioned copolymer has -20 degrees C - good -80 degrees C, and -35 degrees C - its -70 degrees C are desirable. When Tg becomes higher than -20 degree C, flexibility is insufficient and it stops suiting the item of elastic stability (%) of JIS-A5758 (structural sealing material). Moreover, since each copolymerization component of (b), (c), and (d) of high Tg is included, it is difficult for Copolymer Tg to turn under from -80 degree C.
[0016] Tg of the above-mentioned copolymer means the value computed by the formula of following FOX.
[Formula of FOX] 1-/Tg=Wa/Tga+Wb/Tgb+ ... (in the glass transition temperature (K) of a copolymer, Tga and Tgb, and ..., the glass transition temperature of Monomer a, Monomer b, and the homopolymer of ..., Wa and Wb, and ... show [Tg] Monomer a, Monomer b, and the weight fraction of ... among a formula.)
The value according to the specification of Japanese emulsion Semiconductor Equipment & Materials

International can be used for the glass transition temperature of the homopolymer of the monomer used for the above-mentioned count. The value is shown in Table 1. Moreover, according to Japanese emulsion Semiconductor Equipment & Materials International specification, about the copolymerization monomer which cannot check glass transition temperature of a homopolymer, if it is less than 5 % of the weight, it is excludable from count.

[0017]

[Table 1]

略号	化合物名	Tg (℃)
EHA	アクリル酸2-エチルヘキシル	-70
BA	アクリル酸ブチル	-52
EA	アクリル酸エチル	-22
AN	アクリロニトリル	104
AA	アクリル酸	106
MAA	メタクリル酸	185
GMA	メタクリル酸グリシジル	46

[0018] As for the weight average molecular weight of the above-mentioned copolymer, 10,000-1 million are good, and 100,000-700,000 are desirable. If weight average molecular weight turns around 10,000 the bottom, a water resisting property falls and the adhesive property after a water dipping and the elongation engine performance run short. When it becomes higher than 1 million, flexibility is insufficient and it stops moreover, suiting the item of elastic stability (%) of JIS-A5758 (structural sealing material). In addition, this weight average molecular weight is gel permeation which uses polystyrene as the standard substance. It can measure with a chromatography (GPC).

[0019] The above-mentioned copolymer is obtained by carrying out the emulsion polymerization of each component of above-mentioned (a) - (d). That is, a surfactant is made to distribute each of these components underwater to homogeneity. And a copolymer is obtained by adding and carrying out the polymerization of water-soluble persulfate, a hydrogen peroxide, the alpha-cumyl hydroperoxide, etc. as a polymerization catalyst. In this emulsion polymerization, since water is used as a medium, the heat of polymerization can be distributed and a polymerization reaction can be adjusted. This becomes possible to adjust above-mentioned Tg and weight average molecular weight. The copolymer obtained is obtained in the state of the emulsification (emulsion) distributed in water.

[0020] The above-mentioned elastic sealing compound is manufactured by blending a plasticizer, a dispersant, a lightweight bulking agent, a bulking agent, a desiccation regulator, an antifreezing agent, etc. with the above-mentioned copolymer.

[0021] The above-mentioned plasticizer is used in order to improve membrane formation nature, at the same time it makes the above-mentioned copolymer flexible. As an example of a plasticizer, dibutyl phthalate (DBP), dioctyl phthalate (DOP), tricresyl phosphate (TCP), etc. can be raised. The loadings of this plasticizer have good 12 - 35 weight section to the above-mentioned copolymer 100 weight section, and its 20 - 28 weight section is desirable. If [than 35 weight sections] more [a sealing material's flexibility runs short if fewer than 12 weight sections, and], it will become soft too much and elasticity will tend to become inadequate.

[0022] The above-mentioned dispersant helps amalgam decomposition and distribution of a bulking agent, and it is used in order to lower a sealing material's viscosity. As an example of a dispersant, low-molecular-weight polyacrylic acid ammonium salt, a sodium polyacrylate salt, etc. are raised. The loadings of this dispersant have good 0.1 - 5 weight section to the above-mentioned copolymer 100 weight section, and its 0.5 - 3 weight section is desirable. If [than 5 weight sections] more [distribution of a bulking agent may be bad and may be insufficient also for a sealing material's viscosity stability, if fewer than the 0.1 weight section, and], a water resisting property may get worse.

[0023] The above-mentioned lightweight filler is used for the fall of transportation cost, and the purpose of improvement in workability. As this example, natural light weight aggregate (lapilli), hollow particles, such as a microcapsule, etc. are raised. The loadings of this lightweight bulking agent have good 5 - 20 weight section to the above-mentioned copolymer 100 weight section, and its 7 - 18 weight

section is desirable. If fewer than 5 weight sections, the effectiveness of a consistency fall of a sealing material may be insufficient, the workability at the time of transportation and handling may be inferior, and when [than 20 weight sections] more, there is an inclination for a sealing material's reinforcement to fall.

[0024] The above-mentioned filler raises a nonvolatile matter, there are in eye YASE, i.e., the contraction at the time of desiccation, and they are used in order to make desiccation quick. [few] A calcium carbonate, China clay, talc, etc. are raised as this example. The loadings of this bulking agent have the good 75 - 200 weight section to the above-mentioned copolymer 100 weight section, its 80 - 165 weight section is desirable, and its 90 - 130 weight section is still more desirable. If [than the 200 weight sections] more [the ratio of a pitch may become high and a surface tuck and stain resistance may get worse, if fewer than 75 weight sections, and], a sealing material may become hard and elongation may fall.

[0025] As the above-mentioned desiccation regulator, nonionic surfactants, such as polyoxyethylene alkyl phenyl ether, are used. The loadings of this desiccation regulator have good 0.2 - 4 weight section to the above-mentioned copolymer 100 weight section, and its 0.5 - 3 weight section is desirable. If [than 4 weight sections] more [a sealing material's viscosity stability may become inadequate if fewer than the 0.2 weight section, and], a water resisting property may get worse.

[0026] The above-mentioned antifreezing agent is used in order to reduce freezing initiation temperature. Ethylene glycol etc. is raised as this example. The loadings of this antifreezing agent have good 1 - 7 weight section to the above-mentioned copolymer 100 weight section, and its 1.5 - 5 weight section is desirable. If [than 7 weight sections] more [an antifreeze effect may become inadequate if fewer than 1 weight section, and], a sealing material's water resisting property may get worse.

[0027]

[Example] Invention concerning this elastic sealing compound is explained more to a detail using an example.

[0028] (Example 1)

[Composition of an acrylic emulsion] The temperature up of elemi Norian ES-70 (Sanyo Chemical Industries, Ltd. make) was carried out to the reactor equipped with stirring equipment, the reflux cooling pipe, the thermometer, and the dropping funnel as the water 20.6 weight section and a surfactant at 0.05 weight section preparation and 68 degrees C. Next, it is 1.7 weight ******** about 10% water solution of ammonium persulfate, stirring.

[0029] As a monomer component, moreover, the 2-ethylhexyl acrylate (it abbreviates to "EHA" hereafter.) 88.4 weight section, Butyl acrylate (it abbreviates to "BA" hereafter.) Five weight sections, acrylonitrile (it abbreviates to "AN" hereafter.) Five weight sections, an acrylic acid (it abbreviates to "AA" hereafter.) The 1.5 weight sections and glycidyl methacrylate (it abbreviates to "GMA" hereafter.) With the 0.1 weight sections, as a surfactant component, the elemi Norian ES-70 (Sanyo Chemical Industries, Ltd. make) 2.7 weight section and the noy gene ET-160 (Dai-Ichi Kogyo Seiyaku Co., Ltd. make) 2.1 weight section were emulsified in the water 44.4 weight section, and the monomer emulsion was produced.

[0030] Continuation dropping was carried out having applied the obtained monomer emulsion in the above-mentioned reactor with the dropping funnel for 4 hours. In the meantime, polymerization temperature was kept at 72-75 degrees C, and performed the aging reaction at 75-80 degrees C after dropping termination for 3 hours.

[0031] Reaction mixture is cooled radiationally at 30 degrees C after a reaction. The 25% aqueous ammonia 0.63 weight section, The ADEKANETO B-940 (product [made from Asahi Electrification], defoaming agent) 0.01 weight section, the chloro acetamide (made in [Clariant Japan], preservation-from-decay antifungal agent) 0.46 weight section, and 60% NONIPORU 110 (the Sanyo Chemical Industries, Ltd. make --) The wetting-agent 3 weight section is added and they are 60.1% of nonvolatile matters, viscosity 4650mPas, PH4.8, Tg-62.8 degree C, and weight average molecular weight (it abbreviates to "Mw" hereafter.). The acrylic emulsion of 232,000 was obtained.

[0032] The nonvolatile matter of the obtained acrylic emulsion, viscosity, and pH, Tg and Mw were

measured by the following approach. The result is shown in Table 1.
[0033] According to nonvolatile matter JIS-K6833 (synthetic-resin emulsion test method), 1g of samples was correctly measured to the pan with a diameter [made from aluminum] of 4cm, and it put into the drier kept at 105-110 degrees C, and heated for 3 hours, and the weight after desiccation of a sample was weighed precisely after radiationnal cooling in the desiccator.
[0034] According to viscosity JIS-K6833, about 250g of samples was taken to the poly beaker, it fully agitated with the glass rod after 1-hour immersion in the 25**1-degree C tank, and the viscosity of 20rpm was measured using B mold rotational viscometer.
[0035] pH was measured based on pHJIS-K6833.
[0036] It computed by the formula of FOX of Tg above. In addition, the value according to the specification of Japanese emulsion Semiconductor Equipment & Materials International shown in Table 1 was used for the glass transition temperature of the homopolymer of the monomer used for count. Moreover, according to Japanese emulsion Semiconductor Equipment & Materials International specification, about the copolymerization monomer which cannot check glass transition temperature of a homopolymer, when it was less than 5 % of the weight, it excepted from count.
[0037] Gel permeation which uses Mw polystyrene as the standard substance It measured with the chromatography (GPC,;LA[by Shimadzu Corp.]-6A).
[0038] [Combination of an elastic sealing compound] in the above-mentioned acrylic emulsion 100 weight section As a plasticizer, as the DOP(Daihachi Chemical-industry company make) 25 weight section and a dispersant The low-molecular-weight polyacrylic acid ammonium salt (Sannopuko make; SN DISUPASANTO 5027) 1 weight section, As a lightweight bulking agent, the natural-light-weight-aggregate (lapilli) (product made from SHIRAKKUSUU-B- 02) 17 weight section, As a bulking agent, the calcium-carbonate (Japanese east powdering Make; NS- 100) 100 weight section, The polyoxyethylene-alkyl-phenyl-ether (Sanyo Chemical Industries, Ltd. make; NONIPORU 110) 1 weight section was blended as a desiccation regulator, the ethylene glycol (Mitsubishi Chemical make) 2 weight section was blended as an antifreezing agent, and the elastic sealing compound was manufactured.
[0039] A sealing material's obtained viscosity was measured in rotor #7 and rotational frequency 10rpm using BH mold viscometer (TOKIMEC Make). The result is shown in Table 2. Moreover, according to the following approach, a sealing material's paint film was manufactured and the strong ductility and elastic stability of this were measured. The result is shown in Table 3.
[0040] [A sealing material's paint film production conditions] The polyethylene sheet was stuck on the glass plate, on it, the trowel was used, each sealing material was applied at about 2mm thickness, it exfoliated from the polyethylene sheet after care of health for three days on condition that 20 degrees C and 60%RH, it was recuperated for four days on these care-of-health conditions in the rear face, dry-curing was performed for six days at 50 more degrees C, and the paint film sheet of a test piece was produced.
[0041] [A sealing material's paint film strong ductility Measuring condition] Each sealing paint film produced as mentioned above was pierced and carried out by the dumbbell No. 2, the 20mm marked line was attached to the center section of each sample, and it was used for the strong ductility trial.
[0042] The strong ductility trial used the omnipotent tension tester (Made in Oriental Baldwin, UTM-5T), performed the tension test for between chucks at the rate of speed-of-testing 50 mm/min in 60mm, and measured the elongation and reinforcement between 20mm marked lines of a central part (a trial ambient atmosphere is 26 degrees C and 68%RH).
[0043] From the chart, the elongation (it abbreviates to "SMAX" below.) at the time of the reinforcement at the time of 50% elongation (it abbreviates to "M50" below.) and the maximum reinforcement and its reinforcement (it abbreviates to "TMAX" below.), and the elongation at the time of fracture (it abbreviates to "Sb" below.) were checked, and it considered as the strong ductility measurement result.
[0044] [Elastic restoration sex-test conditions of a sealing material's paint film] Each sealing paint film produced as mentioned above was pierced and carried out by the dumbbell No. 2, the 20mm marked line was attached to the central part of each sample, and it was used for the false trial of elastic stability.

[0045] The false trial of elastic stability expands between 20mm marked lines of a central part to about 32mm. Both ends are fixed, the die length at the time of expanding between the marked lines after 36-hour neglect is measured in 26 degrees C and 68%RH, immobilization is removed, and it is a difference (it abbreviates to "the recovery (1)" hereafter.) with the die length between the marked lines after 1-hour neglect on a Teflon (trademark) plate. And a difference with the die length between the marked lines after 3-hour neglect (it abbreviates to "the recovery (2)" hereafter.) It measured.

[0046] (Examples 2-7, examples 1-4 of a comparison) The acrylic emulsion was produced according to the formula shown in Table 2, the sealing material was manufactured according to the approach of a publication in the example 1, and strong ductility and elastic stability were measured. The result is shown in Table 3.

[0047] Moreover, the commercial sealing material was used for the example 4 of a comparison.

[0048]

[Table 2]

試料番号	試料名	アクリル系エマルジョン										シーリング材粘度
		処方					性状					
		EHA (重量%)	BA (重量%)	AN (重量%)	GMA (重量%)	AA (重量%)	粘度	pH	不揮発分 (%)	Tg (℃)	Mw	
1	実施例1	88.40	5.00	5.00	0.10	1.50	4650	4.8	60.1	-62.8	232,000	57
2	実施例2	88.30	5.00	5.00	0.20	1.50	3900	5	60	-62.7	—	60
3	実施例3	88.20	5.00	5.00	0.30	1.50	4200	4.8	60.3	-62.6	—	57
4	実施例4	88.00	5.00	5.00	0.50	1.50	4050	4.9	60.5	-62.4	—	63
5	実施例5	88.48	5.00	5.00	0.02	1.50	8450	4.8	60.9	-62.8	—	63
6	実施例6	88.45	5.00	5.00	0.05	1.50	3180	5.3	60.8	-62.8	613,000	—
7	実施例7	88.25	5.00	5.00	0.05	1.70	2980	5.3	60.2	-62.6	—	—
8	比較例1	87.75	5.00	5.00	0.75	1.50	3250	5	60.1	-62.2	—	52
9	比較例2	87.50	5.00	5.00	1.00	1.50	2450	5	60.6	-62.0	—	77
10	比較例3	88.50	5.00	5.00	0.00	1.50	3000	5	60	-62.8	1,298,000	49
11	比較例4	-	-	—	—	—	—	—	—	—	—	—

※ アクリル系エマルジョン性状の粘度測定は、BH型回転粘度計，ローター#4，20rpmの粘度測定結果で単位はmPa·sである。
※ シーリング材粘度は測定目盛り表示であり、単位は，×4000mPa·sである。

[0049]

[Table 3]

試料番号	試料名	M_{50}	T_{MAX}	S_{MAX}	Sb	復元率(1)	復元率(2)
1	実施例1	0.75 0.074	0.97 0.095	444%	665%	39.20%	48.70%
2	実施例2	0.85 0.083	1.04 0.102	417%	698%	44.20%	50.70%
3	実施例3	0.92 0.09	1.16 0.114	513%	633%	54.50%	64.60%
4	実施例4	1.16 0.113	1.48 0.145	119%	478%	53.60%	62.60%
5	実施例5	0.55 0.054	0.68 0.067	167%	680%	20.20%	26.50%
6	実施例6	0.6 0.059	0.98 0.096	400%	647%	39.40%	47.40%
7	実施例7	0.6 0.059	1.05 0.103	394%	655%	40.40%	43.20%
8	比較例1	0.93 0.091	1.02 0.1	94%	120%	測定不可	測定不可
9	比較例2	1.49 0.146	1.5 0.147	59%	72%	測定不可	測定不可
10	比較例3	0.56 0.055	0.61 0.06	101%	658%	15.20%	20.20%
11	比較例4	0.55 0.054	0.75 0.074	190%	640%	19.50%	26.00%

※ M_{50}、T_{MAX}の単位は上段がkgf/cm^2，下段がN/mm^2である。

[0050] A result M50, TMAX, SMAX, Sb, the recovery (1), and the recovery (2) are compared with the example 4 (commercial item) of a comparison. Each item (especially recovery (1), recovery (2))

estimates that it is powerful, so that the numeric value is higher than the example 4 of a comparison.
[0051] As for examples 1, 2, 6, and 7, in all items, the numeric value is high rather than the example 4 of a comparison. Therefore, a sealing material's film (recovery was included) physical properties will be excellent in these cases.
[0052] At the example 5 with few amounts of GMA copolymerization than the above, Sb, the recovery (1), and the recovery (2) are TMAX, although it is higher than a criteria article. SMAX It is low.
[0053] Moreover, at the example 4 with more amounts of GMA copolymerization than the above, the recovery (1) and the recovery (2) are SMAX, although it is higher than a criteria article. Sb is lower than the example 4 of a comparison.
[0054] Therefore, with the amount of copolymerization of GMA, a little, the item which falls from the example 4 of a comparison is altogether excellent, and can call the recovery (1) and recovery (2) of a certain thing the thing superior to the example 4 of a comparison as a whole.
[0055] on the other hand -- the example 3 of a comparison which does not copolymerize GMA -- TMAX SMAX not only -- the recovery (1) and the recovery (2) have brought a result lower than a criteria article.
[0056] Furthermore in more examples 1 of a comparison and examples 2 of a comparison of the amount of GMA copolymerization, the recovery was the result of the ability not measuring (elongation required for a trial is insufficient).
[0057] Therefore, it became clear about GMA that the acrylic emulsion mold elastic sealing compound in which film (recovery was included) physical properties are excellent is obtained by copolymerizing 0.05 - 0.2 % of the weight preferably 0.02 to 0.5% of the weight.
[0058]
[Effect of the Invention] The elastic sealing compound by this invention is what was excellent also in the miscibility with a filler, and was excellent in elastic stability by carrying out copolymerization of the little ethylene nature partial saturation polyfunctional cross-linking monomer.

[Translation done.]